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Crystal field and spectrum of Pr^{4+} in $BaPrO_3$

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Abstract

We have measured the absorption spectrum of $BaPrO_3$ in the spectral range 1900–11000 cm^{-1}. All the energy levels of the 4f electronic configuration of the Pr^{4+} ion in a crystal have been observed for the first time. We describe the total set of electron-nuclear states and the integral intensities of the magnetic dipole transitions of the Pr^{4+} in $BaPrO_3$ in the framework of the crystal field approximation. The crystal field parameters corresponding to the real orthorhombic symmetry of $BaPrO_3$ are obtained with the exchange charge model.

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1. Introduction

One of the most challenging problems in the modern physics of lanthanide and actinide compounds is the mechanism of the strong superexchange interaction which gives rise to a long-range magnetic ordering at relatively high temperatures in different oxides, in particular, containing tetravalent f-ions [1]. To treat this problem one needs a detailed knowledge of the f-electron energy levels in the crystal field. Among the lanthanides, only cerium, praseodymium and terbium can be obtained in the tetravalent state, and the case of the Pr^{4+} ion, with only one electron in the 4f shell, is the most promising for experimental and theoretical studies.

The first observation of the optical spectrum of Pr^{4+} in crystals, namely in $BaPrO_3$, was reported in Ref. [2]. In this orthorhombically distorted perovskite (space group Pbnm) praseodymium is tetravalent by valence arguments. The Pr^{4+} ions have an almost octahedral arrangement of six near-neighbour oxygen ions [3]. Deformations and mutual rotations of these octahedra result in a drop of the point symmetry of the fourfold 4*b* position for Pr^{4+} ion in the lattice from O_h to C_i, but a consideration of the spectral and magnetic properties with a reference to the cubic symmetry has been useful as a first approximation [1,2,4]. The Γ_7 ground Kramers doublet and four excited levels (Γ_8, Γ_7', Γ_8' and Γ_6 in the order of growing energies) result from the $4f^1$ electron configuration of Pr^{4+} in the crystal field of O_h symmetry. The quadruplet (Γ_8 and Γ_8') levels split into two Kramers doublets for a crystal field of lower symmetry. Only magnetic dipole optical transitions are allowed, the $\Gamma_7 \to \Gamma_6$ transition being forbidden.

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